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Photocatalyzed Cascade Oxidative Annulation of Propargylamines and Phosphine Oxides

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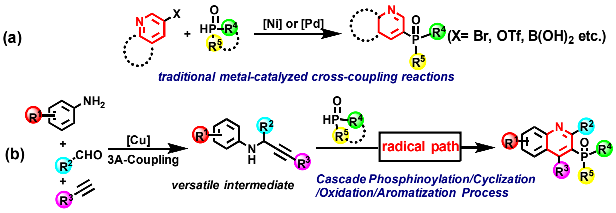
Zheng-Guang Wu[a], Xiao Liang[b], Jie Zhou[a], Lei Yu[a], Yi Wang[a,*], You-Xuan Zheng[a,*], Yu-Feng Li[b,*], Jing-Lin Zuo[a], and Yi Pan[a]

On account of broad utilities of organophosphorous compounds, development of highly efficient and concise phosphination methods is significantly important and urgent. Herein, we disclose a novel method for the synthesis of phosphorylated heterocycles: versatile intermediate propargylamine serving as a new type of radical acceptors incorporating with P-radical via a photocatalytic strategy. This reaction proceeds through a cascade phosphinoylation/cyclization/oxidation/aromatization pathway using readily available starting materials under mild conditions of light with excellent atom economy, catalyzed by AgOAc or fac-Ir(ppy)$_3$. One of the phosphorylated quinolines was selected as an example as an electron-transporting material for fabricating phosphorescence organic light-emitting diodes displaying excellent electroluminescence performances with a maximum external quantum efficiency of 21.9% with negligible efficiency roll-off ratios.

Development of phosphorus chemistry is a subject of continuous interest and great importance because organophosphorous compounds have a wide range of applications in organic chemistry as ligands for transition metal catalysts, in medicinal science as biologically active molecules, as well as in material chemistry as π-conjugated materials.[1] Due to the introduction of phosphorus atom into organic molecules can induce marked changes in their physical and chemical properties, phosphination has become a prevalent and powerful strategy in the design of new molecules. In light of this, numerous efforts have been devoted to developing effective phosphination methods. The transition metal-catalyzed cross-coupling reactions are the traditional strategies for C−P bond formation (**Scheme 1a**).[2] Recently, with the rapid progress of radical chemistry, alternative methods utilizing the active phosphoryl radical [R^{1}R^{2}P-(O)·] to construct the C–P bond have been explored. In fact, the P−H bond of phosphine oxides tends to undergo hemolytic cleavage process *via* the mediation of Ag(I), Mn(III), or peroxides to generate P-centered radicals. Inspired by this elegant reactivity, various methods to incorporate a phosphoryl group with radical acceptors such as internal alkynes, alkynoates, isocyanides, and *N*-arylacrylamides to synthesize phosphorus-containing compounds have been established.[3]

Propargylamines are synthetically versatile intermediates for the construction of biologically active heterocycles, which can be easily prepared *via* 3A-coupling of amines, aldehydes and alkynes, like “bread and butters” serving as the most basic components for achieving a series of cascade reactions with diversity and complexity.[4] We presumed that propargylamine could be employed as a novel radical acceptor to incorporate with P-centered radicals for phosphorylated quinolone synthesis (**Scheme 1b**). The electronic nature of such intermediate defines the chemical selectivity of radical-induced cyclization. The active vinyl phosphoryl species would only attack the *ortho*-positon of the arylamines, forming the kinetically favored six-membered ring. This strategy would benefit from simple raw materials, diverse structures and excellent atom economy compared with the traditional metal-catalyzed coupling methods. Furthermore, the phosphorylated quinoline derivatives could be applied as novel electron-transporting material (ETM) for fabricating organic light-emitting diodes (OLEDs) which might expand the utility of this phosphoylation methodology.



Scheme 1 Radical Cascade Reaction for the Synthesis of Phosphorylated Heterocycles.

Firstly, a variety of silver salts were explored for this reaction of propargylamine **1a** and diphenylphosphine oxide **2a** (**Table S1**). To our delight, the phosphorylated quinoline could be obtained smoothly in 76% yield with AgOAc (3.0 eqiv) as the

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[a] *State Key Laboratory of Coordination Chemistry, Jiangsu Key Laboratory of Advanced Organic Materials, Collaborative Innovation Center of Advanced Microstructures, School of Chemistry and Chemical Engineering, Nanjing University, Nanjing, 210023, P. R. China.*

[b] *College of Chemistry and Molecular Engineering, Nanjing Tech University, 210009, P. R. China.*

Email: yiwang@nju.edu.cn, yxzheng@nju.edu.cn, yufengli@njtech.edu.cn

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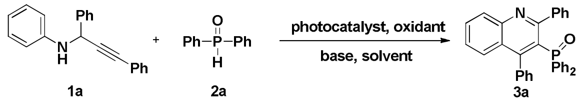
promoter through a cascade radical process. Nevertheless, this transformation required an excess amount of AgOAc and high temperature. Therefore, an effective, mild and ecofriendly phosphorylation strategy is highly desired.

Table 1 Optimisation of the reaction conditions[a]



entry	catalyst	oxidant	base	solvent	yield[b]
1	$Ru(bpy)_3{\cdot}6H_2O$	PIDA	-	DMF	trace
2	$(dfppy)_2Ir(dtbpy)PF_6$	PIDA	-	DMF	13%
3	$Ir(mppy)_3$	PIDA	-	DMF	44%
4	$Ir(ppy)_3$	PIDA	-	DMF	53%
5	FIrpic	PIDA	-	DMF	37%
6	EosinY	PIDA	-	DMF	15%
7	$Ir(ppy)_3$	PIDA	Na_2HPO_4	DMF	50%
8	$Ir(ppy)_3$	PIDA	$NaHCO_3$	DMF	62%
9	$Ir(ppy)_3$	PIDA	LiOAc	DMF	42%
10	$Ir(ppy)_3$	PIDA	K_2CO_3	DMF	31%
11	$Ir(ppy)_3$	PIDA	DABCO	DMF	trace
12	$Ir(ppy)_3$	PIFA	$NaHCO_3$	DMF	trace
13	$Ir(ppy)_3$	$Ph_2I^+OTf^-$	$NaHCO_3$	DMF	32%
14	$Ir(ppy)_3$	TBHP	$NaHCO_3$	DMF	trace
15	$Ir(ppy)_3$	BQ	$NaHCO_3$	DMF	NR
16	$Ir(ppy)_3$	CBr_4	$NaHCO_3$	DMF	NR
17	$Ir(ppy)_3$	PIDA	$NaHCO_3$	DMSO	40%
18	$Ir(ppy)_3$	PIDA	$NaHCO_3$	NMP	45%
19	$Ir(ppy)_3$	PIDA	$NaHCO_3$	CH_3CN	15%
20	$Ir(ppy)_3$	PIDA	$NaHCO_3$	acetone	trace
21[c]	$Ir(ppy)_3$	PIDA	$NaHCO_3$	DMF	NR
22[d]	**$Ir(ppy)_3$**	**PIDA**	**$NaHCO_3$**	**DMF**	**78%**
23[e]	$Ir(ppy)_3$	PIDA	$NaHCO_3$	DMF	0

[a] Reaction condition: propargylamine (0.3 mmol), diphenylphosphine oxide (0.6 mmol), photocatalyst (3 mol%), oxidant (0.6 mmol), base (0.45 mmol), solvent (3.0 mL), under an ambient atmosphere for 36 h. [b] Isolated yield. [c] In dark. [d] Under argon. [e] Under oxygen. PIDA = phenyliodine diacetate. BQ = 1,4-benzoquinone.

In recent years, visible-light-induced photoredox catalysis has drawn great attention due to the inherent green character of light, high reactivity, and good functional group tolerance. Several recent studies have revealed that the diarylphosphine oxides could undergo single electron transfer (SET) process to afford P-centered radical by a reductive quenching cycle of the excited state of photocatalyst.[5] Inspired by this, a new pathway employing visible-light-induced reaction of propargylamines and phosphine oxides for organophosphorous compounds was investigated.

Our study started with a model reaction of propargylamine **1a** with diphenylphosphine oxide **2a** in the presence of various photocatalysts and oxidants (**Table 1**, and **Table S2**). A catalytic amount of ruthenium bipyridine complex and 2.0 equivalent of PIDA gave trace product after stirring in DMF under ambient atmosphere for 36 h (Table 1, entry 1). When using iridium complexes or Eosin Y, the desired phosphorylated product was formed in low to moderate yields (entries 2-6). Among them, $Ir(ppy)_3$ exhibited the best catalytic effects with the yield over 50% (entry 4). Fortunately, after screening the various bases (entries 7-11), $NaHCO_3$ could promote the reaction yield up to 62%. An investigation of solvents demonstrated that DMF was the optimal choice with the yield of 62%, which was better than other solvents, such as DMSO (40%), NMP (45%), CH_3CN (15%), acetone (trace). The reaction efficiency was further enhanced up to 78% when performed under argon atmosphere (entry 22). No product was observed under oxygen or in darkness (entries 21, 23). Furthermore, a small proportion of propargylamine were oxidized to propargylimine, and the side reaction leads to the insufficient yield of phosphoryl radical cyclization.

With the optimized reaction conditions, the generality of the method was investigated and the products were summarized in **Scheme 2**. First, the scope of the reaction was explored with respect to the propargylamine component. As shown in **Scheme 2**, various functional groups on the aromatic rings of Ar1, Ar2 and Ar3 displayed good tolerance under the optimized conditions. Propargylamines with whether electron-donating (methyl, methoxyl and *tert*-butyl) or electron-withdrawing (fluorine, chlorine and trifluoromethyl) groups can react smoothly with diphenylphosphine oxides to give the corresponding 3-phosphorylated quinolines in moderate to good yields. In addition, the propargylamines with bulk naphthyl and biphenyl group at the aromatic rings of Ar1 and Ar2 could also be transferred to give **3e** and **3g** with 60% yield. Furthermore, substrate with other heteroarene such as thienyl could also react efficiently with **2a** to afford the expected product **3o** in 33% yield. For the scope of phosphine oxide derivatives, various P(O)−H compounds were employed to react with **1a**, giving the corresponding products **3p**−**3r** in 47−60% yields. Notably, the phosphaphenanthrene derivative could also generate the P-radical to furnish the 3-cyclophosphate quinoline **3q**, which was reported first time. Furthermore, the above mentioned reactions can also be performed in one-pot four-component process with corresponding amines, aldehydes, alkynes and phosphine oxides (**Scheme S1**).

Based on the above experimental results in combination with literatures,[5b,5d] a plausible mechanism was depicted in **Scheme 3**. The photocatalyst Ir(III) is activated by visible light to the excited-state Ir(III)*, which was oxidized by PIDA to Ir(IV). The high valence of Ir(IV) would oxidize diphenylphosphine oxide to produce Ir(III) and P-radical cation. Thereafter, the key intermediate P-centered radical would be generated through a hydrogen-atom-transfer (HAT) process, which then undergoes intermolecular addition with radical acceptor **1a** to form the alkenyl radical **4**. Subsequently, intramolecular cyclization of **4** produces a cyclohexadienyl-type radical intermediate **5**, which goes through a single electron oxidation transfer process to

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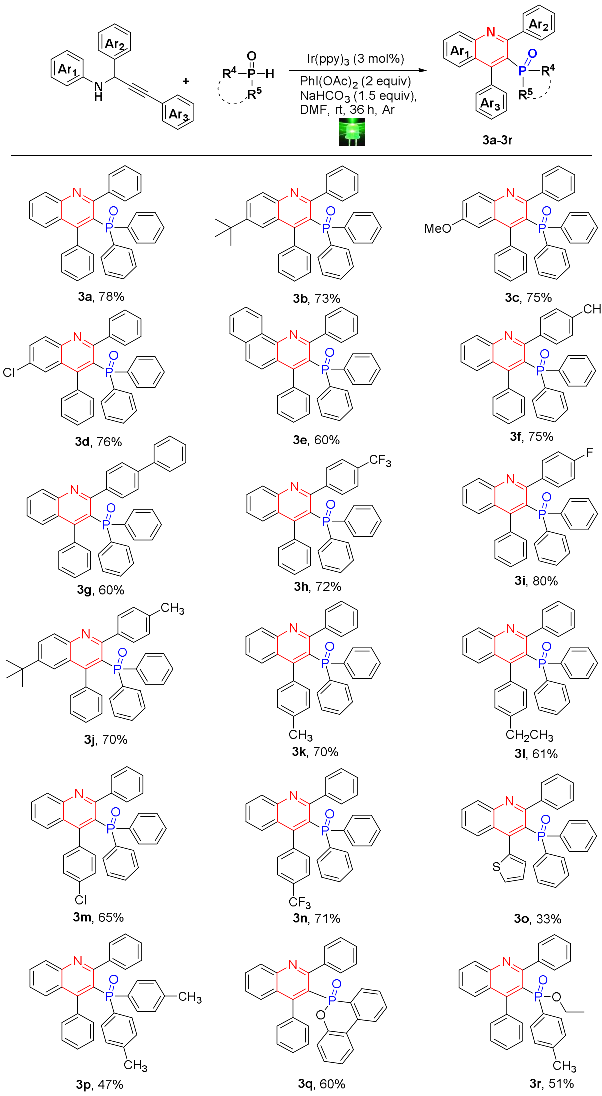
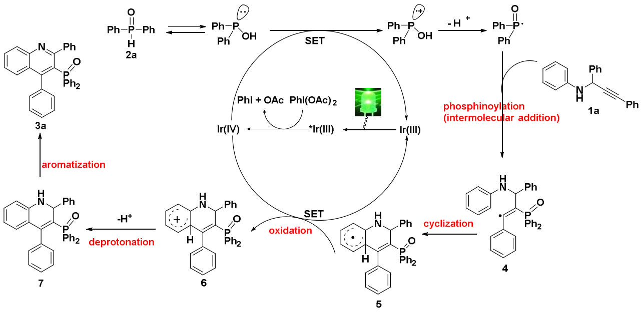


generate cation intermediate **6**. Finally, tandem deprotonation of **6** and aromatization of **7** afford the target product **3a**.

Scheme 2. Scope of the Reaction of Propargylamines with Phosphine Oxides.

In recent decades, phosphorus-containing heterocycles have been widely used as electron-accepting building blocks in materials for organic light-emitting diodes (OLEDs). In virtue of excellent electron transport property, the phosphoryl (P=O) moiety has been extensively introduced in phosphorescent metal complexes,[6] bipolar host and electron-transporting materials (ETM) .[7] In this case, we selected compound **3a** ((2,4-diphenylquinolin-3-yl)diphenylphosphine oxide: QDPO) as an example to be employed as an efficient ETM for OLEDs. First, the physical properties of QDPO, including UV–vis absorption, photoluminescence spectra and thermal stability, were measured, and the key spectral are presented in **Figure S1** and the data are summarized in **Table S3**. The LUMO/HOMO energy levels of QDPO were estimated as 2.80 and 6.65 eV, respectively (**Figure S2 and Table S3**).

Scheme 3 Plausible Mechanism for Photocatalytic Phosphorylation.



To assess the performances of QDPO as the ETM, a typical OLED device was fabricated using widely reported materials with the configuration of indium tin oxide (ITO)/MoO_3 (5 nm)/TAPC (di-[4-(*N*,*N*-ditolyl-amino)-phenyl]cyclohexane) (30 nm)/ $Ir(tfmppy)_2tpip$ (8 wt%) : mCP [1,3-bis(N-carbazolyl)benzene] (10 nm)/QDPO (30 nm)/LiF (1 nm)/Al (100 nm). In this device, MoO_3 and LiF served as hole- and electron-injecting interface modified materials, respectively. TAPC and mCP acted as hole-transporting and host material, respectively. The $Ir(tfmppy)_2tpip$ was also developed by our group as a green emitter containing P=O bonds in ancillary ligand.[6a] The chemical structures of the used materials as well as the energy level diagram of the device architecture were illustrated in **Figure S4**. In order to compare the device performances with conventional materials, the other device using typical electron-transporting material TPBi (2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-*H*-benzimidazole)) with similar configuration was also fabricated.

The current density – voltage – luminance, current efficiency – luminance, power efficiency - luminance and external quantum efficiency –luminance curves of the devices are presented in **Figure 1** and **Figure S5,** and the performance data are summarized in **Table 2**. Device based on QDPO exhibits a lower turn-on voltage of 2.9 V than that of TPBi-based device (3.1 V), possibly due to the lower LUMO level and high electron mobility of QDPO. Furthermore, excellent EL performances with the maximum luminance close to 40000 cd/m^2, the peak current efficiency ($\eta_{c,max}$) of 83.53 cd/A, a maximum external quantum efficiency (EQE_{max}) of 21.9% and a peak power efficiency ($\eta_{p,max}$) of 67.27 lm/W were achieved by device using QDPO. These performances are higher than those of the TPBi-based device, which showed a $\eta_{c,max}$ of 64.00 cd/A, EQE_{max} of 16.7% and a $\eta_{p,max}$ of 49.07 lm/W (**Table 2**). In general, because of the relative long excited state life-time of the Ir(III) complex, phosphorescent OLEDs exhibit severe roll-off at a higher current density due to triplet–triplet annihilation and triplet-polaron annihilation. Noticeably, with increase of the current density, the device exhibits small efficiency roll-off ratios with the η_c of 79.36 and 82.42 cd/A at practical brightness of 100 cd/m^2 and 1000 cd/m^2, respectively, as well as EQE of 20.8% and 21.6% without any light-outcoupling technic. The electroluminescence spectra of both devices reveals the emission maxima at 523 nm with the corresponding Commission Internationale de l'Éclairage (CIE) color coordinates of (0.32, 0.64) and no emission from host and adjacent materials (**Figure 1a**). This results indicate that the recombination of the carriers (hole and electron) occurs only in

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Table 2. Key EL Data of the OLEDs.

Device	$V_{\text{turn-on}}$[a)] [V]	$L_{\max}$(voltage)[b)] [cd m^{-2}(V)]	$\eta_{\text{c.max}}$[c)] [cd A^{-1}]	$\eta_{\text{c,100/1000}}$[d)] [cd A^{-1}]	$\eta_{\text{p.max}}$[e)] [lm W^{-1}]	$EQE_{\text{max,100,1000}}$[f)] [%]
QDPO	2.9	38929(13.5)	83.53	79.36/82.42	67.27	21.9/20.8/21.6
TPBi	3.1	40422(13.3)	64.00	60.13/62.70	49.07	16.7/15.7/16.3

[a)]$V_{\text{turn-on}}$: turn-on voltage recorded at a brightness of 1 cd m^{-2}; [b)]$L_{\max}$: maximum luminance; [c)]$\eta_{\text{c,max}}$: maximum current efficiency; [d)]current efficiencies measured at the brightness of 100 cd m^{-2} and 1000 cd m^{-2}; [e)]$\eta_{\text{p,max}}$: maximum power efficiency; [f)]$EQE_{\text{max, 100, 1000}}$: maximum external quantum efficiency, external quantum efficiency at 100 cd m^{-2} and 1000 cd m^{-2}.

the EML, and the completely energy transfer from host to dopant. The outstanding QDPO-based device performances may be attributed to the following facts: 1) the phosphoryl group P=O and pyridine ring would be helpful for electron transporting; 2) the suitable LUMO level will contribute to electron injection and the lower HOMO level benefits for holes/excitons blocking. Therefore, a better balanced charge injection and transport will promote the recombination of electrons and holes and broaden the recombination zone as well as lead to the suppressed current leakage in the devices.

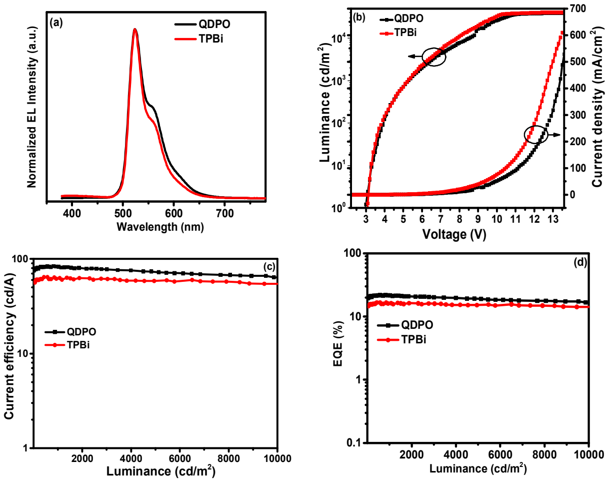

Figure 1. Device characteristics: (**a**) EL spectra at 50 mA. (**b**) current density and luminance versus voltage. (**c**) current efficiency versus luminance. (d) external quantum efficiency versus luminance.

In conclusion, we have developed a simple and effective method for the synthesis of functionalized phosphorylated hereocycles through a cascade phosphorylation/cyclization/ oxidation/aromatization process of propargylamines and phosphine oxides. The novel compound was used as ETM for fabricating OLEDs and excellent EL performances were achieved. We anticipate that this methodology will be particularly useful for the synthesis of organic functional materials with P=O moiety.

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